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Interim Storage of Excess Plutonium:
An Assessment of Options (U)

John M. Haschke
Joseph C. Martz

June 17, 1993

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Interim Storage of Excess Plutonium: An Assessment of Options (U)

by

John M. Haschke and Joseph C. Martz**0. Abstract (U)**

A technical evaluation of options for interim storage of excess weapons-grade plutonium is presented. Storage of plutonium as intact pits, altered pits, and extracted material is evaluated. Alternative technologies within each of these general categories are outlined along with advantages and disadvantages of each specific option. A range of technical issues is examined including feasibility of the concept; maturity of the technology; environmental, safety, and health concerns; nuclear materials accountability; transparency; waste generation; and preservation of flexibility for ultimate disposition. The advantage of each storage option in reducing the risk of direct reuse of the stored material in fabrication of a nuclear weapon is considered. In addition, the ease of reconstitution to a usable form is examined. Although storage as altered pits or extracted material may reduce the likelihood of rapid, immediate rearmament by a nuclear state, none of the options is considered more effective than any other in preventing construction of a nuclear device by a terrorist or subnational group.

We recommend that excess plutonium be stored as intact pits. If extraction of plutonium is required, we urge that material be left as metal for interim storage. If treaty obligations or political considerations forbid storage of pits or plutonium metal, storage of extracted plutonium as oxide is the most preferred storage form. Procedures must be developed to efficiently convert large quantities of metal to certified oxide with minimal waste generation. We suggest that all existing oxide (and oxide-containing residues) be stored in their existing material forms.

If a disablement technique becomes available which does not significantly alter the plutonium form or chemistry, does not introduce additional waste, preserves the pit as a primary storage container, and remains compatible with existing extraction processes, then it should be considered an acceptable option as well.

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1. INTRODUCTION ~~UNCLASSIFIED~~

Recent agreements with Former Soviet Union (FSU) states will result in the removal of approximately 17,000 nuclear weapons from the active stockpile.¹ This development will dramatically alter the nuclear materials economy. In past decades, the weapons complex has been driven by frequent nuclear material shortages. However, the future will be characterized by an excess of plutonium. As such, storage of plutonium from these weapons has become one of the most important challenges for the nuclear weapons establishment. The nuclear materials effort must be redirected from one with emphasis on production and reprocessing of plutonium for stockpile needs to one with emphasis on maintenance of the enduring stockpile and on dismantlement of weapons and management of excess plutonium.²

Interim storage of excess plutonium is complex and includes both technical and political issues. Three general options have been identified for storage of excess material: (1) storage as intact pits, (2) storage as altered pits, and (3) storage in a suitable material form after extraction from the pit.^{3,4} An adequate evaluation of these options must consider technical capability as well as facility requirements and availability. Differences in pit design (e.g., an outer container of stainless steel for certain units and beryllium outer shells for others) must be considered. Generation of radioactive waste is a particularly important component of environmental, health, and safety (ES&H) concerns related to these storage options. Additional issues with strong technical and political implications include rearmament and proliferation concerns. As a result, materials control and accountability (MC&A), transparency, and safeguards and security must also be addressed.

Readiness requirements and the time frame encompassed by "interim storage" are important factors for evaluating technical options. Both durations are somewhat uncertain. Implementation of various technical options within a period as short as 2 years has been initially suggested, while various time frames from 5 to 15 years have been subsequently proposed.⁵ Technical options must also be examined in light of an interim storage period that will likely span several decades and could extend to as much as 100 years.

The focus of this report is a comprehensive technical evaluation of potential storage methods identified in a complex-wide assessment of storage options sponsored by the Department of Energy Arms Control and Nonproliferation office (AN).⁶ As participants in that study, we encountered a wide diversity of opinion regarding the merits and suitability of various storage alternatives. As the coordinators of the DOE-AN study

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found, a comprehensive and internally-consistent report is difficult to prepare from input provided by several contributors with diverse perspectives. A credible report on plutonium storage must consider all pertinent information on physical and chemical properties of materials, the maturity levels of candidate technologies, potential hazards and complications associated with storage options, requirements for certification and packaging, and surveillance and monitoring of stored material. A comprehensive assessment must include relevant information from earlier studies by qualified experts as well as all potential options from diverse sources. Conflicting perspectives must be addressed. For example, non-proliferation advantages cited for certain chemically altered storage forms are inconsistent with a consensus of technical opinion stating that those materials can be used as nuclear explosives and that they are easily reconstituted to plutonium metal. Since our analysis focuses on technical issues, discussions of such topics as the process capability of specific facilities, material control and accountability (MC&A) issues, safeguards and security, and transparency concerns are limited. We attempt to identify and evaluate a range of technical issues associated with various options proposed for interim storage. Such an evaluation of storage forms cannot be divorced from the procedures and conditions required to obtain and certify those forms or from the procedures and conditions required to reconstitute the plutonium into a form suitable for fabricating a nuclear device.

2. DESCRIPTION OF STORAGE OPTIONS

2.1 Storage as Intact Pits

Storage of intact pits is an approach that employs the intrinsic characteristics that made the pit suitable for extended stockpile storage in a nuclear weapon. For an "intact" configuration, tubes might be coiled and immobilized to prevent torquing motion which could break welds or joints.⁷ Although the title of this storage category seems to preclude any alteration of the pit, the possibility of removing tubulation has been suggested as a procedure to facilitate packaging. Additional storage options appear in the packaging configuration. Issues include the number and specifications for barriers between the pit and the ambient environment, and whether pit containers are stored within insulated shipping containers.

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2.2 Storage as Altered Pits

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remains as a containment vessel. Two levels of alteration are recognized: (1) denying direct reuse of the pit in a nuclear device by mechanical or physical alteration, and (2) altering the utility of the nuclear material by chemical methods that make reconstitution (recovery and reprocessing) of the plutonium prohibitively difficult.

2.2.1 Mechanical Methods.

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2.2.2 Physical Methods.

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2.2.3 Chemical Methods.

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2.3 Storage as Extracted Material

Storage of extracted material is considered attractive by some because of the potential to destroy the pit configuration and prevent easy reconstitution. The variety of candidate forms considered for interim storage include metals (unalloyed and alloyed), plutonium compounds, and mixtures. In all cases, the pit is bisected to gain access to plutonium. Extraction processes are used to separate the plutonium from other pit components prior to processing to the desired material form.

2.3.1 Chemical Options. In certain options, plutonium is stored as unalloyed metal.

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An alternative extraction process for production of dioxide or metal is based on aqueous methods. Plutonium compounds or mixtures are dissolved in aqueous mineral acid (HNO_3) and plutonium is separated from other elements by ion exchange or solvent extraction.^{20,21} Oxide is obtained by precipitating an insoluble plutonium compound (e.g. the hydrated oxalate, $\text{Pu}(\text{C}_2\text{O}_4)_2 \cdot 6\text{H}_2\text{O}$) which is subsequently decomposed at high temperatures (calcined). Metallic plutonium is prepared by reaction of oxide with calcium metal at temperatures above 850 °C.

Plutonium in aqueous solution is not considered a viable storage form. Issues of chemical and radiolytic compatibility, long-term stability, criticality safety, storage volume, configuration requirements, surveillance, and accountability activities all suggest that storage of material as an aqueous plutonium solution is unacceptable. Detailed justification of this decision is beyond the scope of this report, but interested readers are directed to the extensive body of literature on properties of plutonium in solution.²²

2.3.2 Dilution Options. Certain alternative storage forms for extracted plutonium are obtained by dilution of weapons-grade material. Options include addition of plutonium isotopes other than ^{239}Pu , dilution with highly radioactive fission products or high-level waste (HLW), and dilution by mixing with non-nuclear materials such as glass or sand (e.g., vitrification).

3. TECHNICAL EVALUATION OF STORAGE OPTIONS

Technical issues related to the three primary storage options are sufficiently diverse that each method is considered separately. Reconstitution of material from various candidate forms, waste generation, and transparency issues are general areas that are addressed as individual topics in section 4.

3.1 Storage as Intact Pits

Since all pit types have been in stockpile storage for some period of time within a weapon configuration, difficulties are not anticipated during continued storage as disassembled but intact units. If the pit is unaltered, technical concerns are limited to those already encountered during surveillance of units from stockpile.²³ Certain requirements imposed for storage in stockpile remain during interim storage of pits after weapon disassembly. Concerns and constraints are encountered both in the configuration of the storage container and in establishment of storage conditions. The number of positive closures required to establish confidence that plutonium is contained must be determined.

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The temperature of plutonium components in storage is an important parameter that is strongly influenced by the self-heating properties of plutonium.²⁴

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Stringent constraints on storage temperature are anticipated for pits retained for possible reuse in weapon manufacture. Confidence in the integrity of reuse pits would be compromised if temperatures exceeded the 65–70°C range for storage of stockpile weapons. This constraint reflects a limited experience with long-term storage of plutonium metal outside of this temperature range. Problems with unknown aging effects in reuse pits would be compounded by high-temperature storage.

Storage temperature is not the only constraint for intact pit storage. The potential for plutonium release during an accident in transport or storage is of foremost concern. Plutonium in pits is potentially released by a number of mechanisms: mechanical breach, low-melting eutectic formation, and atmospheric oxidation. Transport and storage containers are designed to preserve structural integrity to the pit and prevent release by mechanical breach. Mitigation of other release hazards requires a different approach. A fire that produces temperatures in excess of the plutonium melting point (640°C) poses a hazard for release and dispersal of nuclear material.

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All pits to be removed from

stockpile have ultimate containment layers of either beryllium or steel.

Other pit-specific storage requirements must be considered. In addition to differences in metallurgical phase, plutonium content, heat generation, and outer confinement material, important pit features include size, number and position of tubes, and the possible inclusion of neutron generators containing tritium or ^{238}Pu . Each of these variables influences the packaging configuration. For example, fixtures for constraining pit and tube movement and for dissipating heat during storage are required for each of the 10-15 pit types in the dismantlement schedule. Likewise, development of individualized surveillance programs is essential for establishing the suitability of each pit type for continued storage and for identifying those types requiring remedial action or disassembly.

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3.2 Storage as Altered Pits

Effectiveness of alteration or deformation as a denial method can only be determined on the basis of yield calculations for each deformed pit. The level of yield reduction must be considered if denial is the ultimate goal

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3.2.1 Mechanical Methods. In mechanical alteration, the pit serves as a containment vessel after deformation or dudding procedures. Concerns regarding packaging requirements, temperature limits, and fire scenarios are similar to those outlined for intact pits. Additional concerns include maturity and general applicability of the technologies, recertification of the pit as a container, and effectiveness of the denial process. Although simple in concept, mechanical deformation methods are not extensively developed and generally applicability to all pit types is uncertain.

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Regardless of the method of mechanical denial, an appropriate level of surveillance is necessary to ensure long-term stability and containment of plutonium.

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particular concern for dudding masses because of potential difficulties in welding or pinching due to residues which might remain on tube walls after injection of material.

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3.2.2 Physical Methods.

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As with mechanical denial methods, specific procedures must be developed for each pit type.

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3.2.3 Chemical Methods. Technical issues related to injection of reactive materials are extensive. Individual consideration of chemical options is necessary because of the broad diversity in behavior for candidate processes. In addition to an increase in material volume during reaction, the rate and heat of reaction for chemical disablement processes are particularly important parameters. General statements about chemical methods are limited in number. Certain concerns are shared with other disablement methods.

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Some have suggested that hydride intermediates might spontaneously form the desired alloy at low temperatures. This seems doubtful. Extensive experiments conducted with freshly prepared plutonium hydride and plutonium metal powders in thin (0.25 mm thick) Al foil containers at temperatures up to 500°C show no evidence of alloying.⁵⁹ A possible explanation for this observation is provided by thermodynamic data for the hydride⁶⁰ and for intermetallic compounds of plutonium.⁶¹ Although enthalpies and free energies of formation are not available for the specific alloys of interest, values reported for compounds of other elements (e.g., Fe, Rh, Ru, Pt, etc.) suggest that Al and Cu will not react with PuH_x because of unfavorable thermodynamics (i.e., PuH_x is more stable than the alloys) or will occur with production of insufficient amounts of heat to drive a self-sustained alloying process. These results are consistent with unsuccessful efforts to initiate reaction between PuH_x and low-melting-point metals.⁶²

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Although conversion of plutonium metal to other chemical forms reduces certain ES&H concerns (e.g., corrosion of steel containers during a fire), the risk of dispersing plutonium into the environment may be increased by chemical alteration.

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3.3 Storage as Extracted Material

Flexibility in storage options is increased if plutonium is removed from the pit containment vessel. Restrictions on storage temperature are less stringent because material expansion can be accommodated and fire resistance can be incorporated in container design. Difficulties and constraints associated with *in situ* formation of compounds (hydride, oxide, nitride, alloys, etc.) are significantly reduced by not relying on the pit container to serve as a reaction vessel. However, extraction of plutonium from the pit allows material to more readily contact other chemical species in the process environment. As discussed in sections 3.3.2 and 3.3.4, the potential consequences of such exposures are strongly influenced by the physical properties (such as surface area) of the storage form. Since a technical evaluation related to storage of extracted material is most conveniently addressed by focusing on issues instead of material form, that format is adopted for this section.

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3.3.1 Chemical Reactivity. The chemical reactivity of a candidate form is an important consideration. Unfortunately, chemical reactivity is often the preeminent (and only) issue considered in evaluating the suitability of a material for storage.⁶⁸ An assessment of reactivity for a material must consider both thermodynamic and kinetic concerns in the specific storage environment. Plutonium oxide is the most stable form of plutonium in air and other oxygen-rich media.⁶⁹ Metal and all other plutonium-containing compounds except fluorides (PuF_3 , PuF_4 and PuF_6) are unstable relative to oxide in air. Even under aggressively inert conditions, the surfaces of all candidate materials are covered by oxide films. Although PuO_2 is more stable than other potential storage forms and is considered to be the equilibrium oxide phase in air, recent results for the reaction of water vapor indicate the presence of a thermodynamically-favored, higher-valent oxide, a mixed-valent compound with the $\text{Pu(IV)}_4\text{Pu(VI)}\text{O}_{11}$ composition ($\text{PuO}_{2.2}$).⁷⁰

Oxidation reactions of candidate storage forms exhibit a broad spectrum of kinetic behavior. Massive plutonium metal (> 0.5 mm thick) reacts slowly with oxygen and is routinely handled in air without risk of rapid reaction. Spontaneous ignition is only observed if the metal is in a finely divided (< 0.2 mm thick) state and if the temperature also exceeds 150°C .⁷¹ In comparison, plutonium hydride is pyrophoric and reacts violently upon exposure to air at room temperature.⁷² Nitride and carbide also spontaneously react with air and moisture.⁷³ Chemical reactivities of alloys are difficult to evaluate because information on most alloy oxidation kinetics are unknown. A particularly important reaction of oxide and oxide-coated materials is the rapid and tenacious sorption of water from the atmosphere;⁷⁴ several monolayers of water remain irreversibly bound at 100°C in high vacuum.^{75,76,77}

Several reactions of plutonium and its compounds are of interest because of their potential ES&H impact. As noted in Section 3.1, corrosion of stainless steel by molten Pu poses a credible hazard because of the potential for plutonium release to the environment. Recall that thermal decomposition of hydride produces corrosive liquid metal and high hydrogen pressures. Loss of containment may also occur with oxide powders during a fire or extended storage due to generation of high pressures from adsorbed species.⁷⁸ These and other pressurization processes are discussed in Section 3.3.5. Plutonium metal provides a measure of inherent protection from pressurization because of its ability to getter reactive surface species without forming secondary gases such as H_2 , N_2 , or C_2H_2 .

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3.3.2 Radiolytic Reactivity. Radioactive decay of plutonium alters the chemical behavior and makeup of molecular and organic compounds in a storage vessel and may alter the stored material or lead to loss of containment.⁷⁹ Radiolytic chemistry is influenced by particle dimension as well as isotopic composition and age of the plutonium. Powdered samples emit a large fraction of the 5.1 MeV alpha decay particles, the primary initiator of radiolytic reactivity for ²³⁹Pu. Recall the high surface area of finely divided plutonium compounds promotes adsorption of large amounts of water, organic molecules etc. As such, alpha particles from radioactive decay have ample opportunity to interact with adsorbed species in the storage environment. The presence of adsorbates is not limited to fine powders: adsorption is observed for all candidate materials because of facile formation of surface oxide. However, the quantity of adsorbate on massive samples such as alloys or metal is small compared to PuO₂ and other finely divided forms. Dioxide products with specific surface areas in excess of 50 m²/g are common.^{80,81} When exposed to ambient glovebox atmospheres for several hours, the mass of a typical oxide sample may increase by more than 1 % due to water adsorption.⁸²

Interaction of alpha particles with adsorbed species, organic materials (e.g., plastics), or gases in contact with the nuclear material leads to chemical bond cleavage and formation of gaseous products.^{83,84,85} Low-molecular-weight species (e.g. O₂ and N₂) primarily dissociate to form gaseous products (e.g., NO₂), while heavier molecules tend to polymerize. Radiolysis of water generates hydrogen and oxygen;^{86,87} alpha interaction with air produces large percentages of dinitrogen monoxide(N₂O) and nitrogen dioxide (NO₂).⁸⁸ Products formed by radiolysis of plastics and other organic materials depend on their chemical composition and typically include hydrogen.⁸⁹ Radiolytic decomposition of polyvinyl chloride (PVC) apparently forms gaseous hydrogen chloride (HCl).

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In sum, the magnitude of radiolytic effects is influenced by two important properties of the material to be stored: the specific surface area (and the associated quantity of adsorbates), and the ability of the material to getter radiolytic products. In both cases, plutonium metal and alloys are expected to have considerably fewer problems with radiolytic phenomena than more finely divided Pu forms.

3.3.3 Helium Release. Helium formed by alpha decay of ^{239}Pu and other isotopes provides an additional source for pressure generation during storage. Although the process is slow ($t_{1/2} = 2.4 \times 10^4$ years), the quantity of helium may become significant if the storage period is long and the fraction of He released by the solid phase is large. The release behavior of massive metal differs from that of oxide because the diffusion rate of He in metal is relatively slow and hence, He is only able to escape from the near-surface region of the solid. By contrast, diffusion of He in oxide is comparatively rapid and the small particle dimensions allow the ready release of all helium created by alpha decay. Experimental data for both ^{238}Pu and ^{239}Pu show that helium is readily released from oxide^{90,91} but is retained as microscopic bubbles at grain boundaries of Pu metal.⁹²

Calculations based on the alpha decay rate and helium release kinetics of metal and oxide show that He pressures greater than one atmosphere are anticipated for oxide in a typical storage configuration after 100 years.⁹³ By comparison, the pressure rise expected for storage of an equivalent amount of metal is approximately 0.001 atmosphere. Although the He release behavior of massive alloys and non-metallic powders is expected to parallel those of the metal and oxide, experimental data are not available.

3.3.4 Container Pressurization

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3.3.5 Release of Plutonium to the Environment. From both environmental and political perspectives, release of plutonium-containing materials to the environment is a serious potential hazard. The two most likely situations that may lead to release are mechanical breach of a storage container and exposure to a fire. Once plutonium has been released to the environment, further spread of material is of prime concern. As such, the dispersal risks associated with various forms is directly related to the particle size and form of the material.⁹⁹

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In contrast to metal or alloys, relatively large fractions of finely-divided forms are in the dispersible range.¹⁰³ 20–25 mass% of the PuO_2 formed by oxidation of the hydride consists of particles with dimensions less than $10\text{ }\mu\text{m}$,¹⁰⁴ while 100 mass% of the oxide formed by pyrolysis of precipitates (e.g., hydrated plutonium oxalate) from aqueous processing is in this dispersible range.¹⁰⁵

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Upon release of plutonium metal to the ambient environment, the potential for oxidation and subsequent dispersal by aerosolization exists. The oxidation rate of Pu in air at 500°C and above is modest and constant ($0.2\text{ g PuO}_2/\text{cm}^2 \cdot \text{min}$ of metal surface).¹⁰⁷ The oxide product formed at these temperatures is relatively coarse with less than 0.1 mass% in the dispersible range.¹⁰⁸

Although container corrosion may also occur during a fire involving stored hydride, the possibility of container rupture exists for other powdered forms because of increased gas evolution and pressure and decreased container strength at high temperatures. The pressure is a combined effect arising from chemical reaction, radiolytic decomposition, helium release, thermal desorption, and thermal expansion. Rupture is expected to result in the dispersal of a larger fraction of powdered forms than would be released from massive metal or alloys.

On the basis of relative particle size distributions, the quantity of plutonium-containing material released to the environment is 1,000 to 10,000 times greater for finely-divided material forms than for massive metal. The risk of corrosion and container failure are likely reduced for alloy storage, but the potential hazard posed by such storage can not be assessed because information on reaction rates, thermodynamics, and particle distributions are not available.

Remediative actions can be employed to reduce the risks associated with containment of metal and powdered forms during a fire. Application of a ceramic coatings on internal container surfaces (e.g., erbium oxide)¹⁰⁹ or insertion of reactive container liners that alloy and chemically immobilize plutonium as a high-melting compound (e.g., aluminum)¹¹⁰

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would greatly reduce the risk of metal release during a fire. The possibility of pressurization could also be reduced for powdered materials by certification to verify that significant quantities of adsorbates are not present.¹¹¹

Use of vented containers for powder storage must be evaluated in light of reduced container structural integrity and the consequences of radiolysis-enhanced corrosion of the nuclear material or the container. Formation of nitrogen oxides from air in a radiolytic environment produces nitric acid if water is also present.¹¹² Mass measurements for oxide stored in vented stainless-steel containers show a mass increase of 2-3% after several years.¹¹³ Reasons for these changes are not known, though the possibility of container oxidation catalyzed by radiolytically-formed nitric acid is under investigation. Rupture disks which function only on pressurization may alleviate the risk of catastrophic container failure during fires or other accidents while preventing exposure of the nuclear material to external ambient storage environments under normal conditions.

3.3.6. Other Environmental, Safety, and Health Issues. Other ES&H issues related to storage of extracted material include nuclear criticality safety, radiation exposure to personnel, and contamination control. Criticality safety procedures are well established for all candidate storage forms. Primary controls include limiting the quantity of stored material and careful inventory and control of physical storage and locations. As an added precaution, storage of plutonium metal and alloys as castings with critically-safe geometries (e.g., thin-walled rings) can reduce the possibility of a criticality incident by deliberate or accidental violation of safety procedures. Criticality safety for finely-divided forms must rely on administrative controls that establish handling procedures and mass limits.

Procedures for limiting worker radiation exposure and controlling spread of radioactive contamination have been well established in the last 40 years. However, increasingly stringent regulatory requirements are anticipated for both areas, and hence, consideration of advanced material handling technologies to reduce these exposures and risks is worthwhile. The advantage of automation in distancing operators from nuclear material and reducing the likelihood of operator error is widely recognized.¹¹⁴ Castings of metal or alloy are amenable to automated handling, as are powdered materials. Procedures differ for handling finely-divided forms and in some instances are more complex than for

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transfer of discrete metal components. However, care must be taken in automation to ensure high reliability among robotic components because frequent breakdowns in automated equipment would only transfer radiation exposure from operating to maintenance personnel.

Storage of plutonium in a form containing nuclear fission products as radioactive diluants would have an enormous impact on radiation exposure levels in a typical plutonium facility.¹¹⁵ Construction of special remote handling capability would be necessary to accommodate such highly radioactive materials. Numerous vitrification plants are in operation which provide for remote process and handling of highly radioactive waste.

3.3.7. Process Technologies. Requisite processing for storage of extracted materials is contingent on the storage form(s) adopted. Constraints on volume, temperature, gas flow, and throughput that restrict *in situ* chemical disablement can be readily addressed for extracted material with appropriately designed equipment. For example, production of oxide via the $\text{PuH}_2 + \text{O}_2$ reaction is an established production process.¹¹⁶ Constraints on temperature and hydrogen removal are easily accommodated by appropriate design of process equipment. Formation of many intermetallic plutonium compounds has been demonstrated on a kilogram scale though production qualified operations have not been developed.^{117,118}

The level of development activity required to implement storage of extracted material is strongly dependent on the choice of storage form. Methods are fully developed for extended storage of metal;¹¹⁹ only transfer of technology from pit fabrication processes is necessary. Experience with storage of oxide is more limited, but the general criteria (e.g., specification of residual polyanion content, adsorbed water loading, oxide stoichiometry, etc.) for preparation of a certified oxide are known. However, procedures to achieve these criteria have yet to be developed. Requirements and technologies for preparing other candidate storage forms are poorly defined.

3.3.8. Packaging. Although specifics of container design are influenced by the storage form, basic features are similar for all options. In addition to satisfying transportation regulations and providing suitable fire resistance, *the primary container must not include organic or covalently-bound materials that might be exposed to plutonium-containing*

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particles. This includes organic and other covalently bound materials introduced either intentionally (e.g., bagging or o-rings) or unintentionally (e.g., adsorbed water on oxide). A second essential requirement is that all containers be sealed (and certified as such) so that communication with the ambient atmosphere during storage is prevented. An extensive study of failures and accidents involving plutonium storage reveals that one or both of these requirements was not met in all cases examined.

3.3.9 Materials Control and Accountability. MC&A is an important technical issue because certification of the nuclear material quantity is necessary after extraction. Accountability methods typically rely on chemical analysis of "representative" samples and material mass measurements to determine plutonium content. Use of mass measurements alone is unacceptable. For example, the Pu content of "dioxide" samples prepared by precipitation from aqueous solution and calcination may be up to 10% less than the theoretical value for PuO₂.¹²⁰

Product homogeneity is essential for obtaining representative samples and obtaining accurate analytical results. Inhomogeneity is not a severe problem for plutonium metal or for binary compounds (e.g., oxide, nitride, hydride) formed by reaction of metal with pure gases. Products formed by reaction of condensed phases or by pyrolysis of solids (e.g., carbides, intermetallic compounds, some oxides) are difficult to homogenize. The existence of multiple solid phases and slow transport processes in a chemical system introduces a series of kinetic limitations that hinder product equilibration. This results in sample inhomogeneity which leads to difficulty in determining actual plutonium content based on "grab" samples and mass measurements. Calculation of Pu content from calorimetry is not as sensitive to sample inhomogeneity, though calorimetry is not as sensitive or accurate as other assay techniques. Problems with inhomogeneity are expected to be particularly acute for plutonium stored in high-level fission waste. Verification of plutonium mass and detection of plutonium diversion would be difficult (if not impossible) with such storage forms.

Results of nondestructive assay (NDA) methods such as neutron and gamma-ray spectroscopies are also sensitive to the sample matrix. Inhomogeneity may alter the results of these measurements. Heavy elements that alter self-absorption characteristics and light elements that participate in alpha-neutron (α -n) reactions are of particular concern.¹²¹

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4. GENERAL TECHNICAL ISSUES

Certain issues are common among all storage forms and options. Concerns about reuse of material, ease of reconstitution to nuclear weapons, waste generation, preservation of flexibility for ultimate final disposition, verification, and transparency of nuclear material extraction and storage are all considered here. All of these topics merit detailed, indepth evaluation far beyond that given below. As such, our discussion is to be considered cursory and is only intended to highlight the major relevant issues and how they relate to various plutonium storage options.

4.1. Reuse and Reconstitution

The motivation for disablement of pits and chemical alteration of extracted plutonium is in denying direct reuse of components and materials in a nuclear device and in making reconstitution to a usable form prohibitively difficult. A careful examination of reuse and reconstitution issues is needed to adequately assess the merits of implementing disablement or chemical conversion processes. As will be seen below, the primary advantage gained from such processes is political, not technical. While reconstitution and reuse of plutonium in any candidate form is technically simple and well within the capability of a subnational or terrorist group,¹²² the potential political message alteration or disablement sends may hold considerable value (i.e., the ability to say we have "done something" to our nuclear material such that it is no longer a "nuclear weapon").

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Further, the benefits of chemically altering pits or extracted material must be weighed against the possibility of direct reuse in device construction and the possibility of conversion of the plutonium form to one more suitable for weapon fabrication.

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DOE b(3) Ironically, facility requirements are likely not an issue for subnational or terrorist groups because ES&H concerns would probably be ignored. In the absence of sophisticated protective equipment, inexpensive protective clothing and particle respirators could be used. The very concerns that might make reprocessing of plutonium expensive and time-consuming for the United States will likely be dismissed by motivated terrorist groups.

Processes for reconstitution of chemically altered plutonium forms utilize a common and adaptable technology base.

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[Recovery of metal from hydride is trivial: since the hydride is thermally decomposed to metal and hydrogen, Pu is obtained by placing hydride in the crucible and heating to 700°C in dynamic vacuum or under a flow of inert gas.¹³¹ Conversion of oxide to metal is also a simple, one-step process.¹³² PuO₂ and a stoichiometric quantity of calcium (Ca) metal are placed in the crucible and the system is closed; the spontaneous reaction between PuO₂ and Ca to form Pu metal and CaO is initiated by heating above 850°C. The process can be accomplished without evacuation because oxygen in the sealed reaction chamber will react with Ca in preference to Pu.

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Advantages of extreme dilution must be weighed against the costs and the storage volume necessitated by the large quantity of material.

4.2. Waste Generation

The level of waste generation is a major consideration in assessing storage options for extracted plutonium. Though advantage is gained by upgrading process technologies, the greatest benefit is achieved by avoiding all unnecessary processing.¹³⁹ The simplest method of limiting waste generation is intuitive: leave materials in their existing form. Interconversion of material forms should be minimized to avoid waste generation and other ES&H concerns associated with material handling. However, potential problems with a particular stored material may require processing to an alternate form to alleviate concerns. Nearly all plutonium in US inventories is currently metal or oxide, with a lesser fraction stored mostly as process wastes including incinerator ash, chloride-rich salts, and aluminum-magnesium compounds (scrub alloy). The inventory existing as carbide, nitride, or hydride is small.

Certain options might be suggested to remediate anticipated storage problems. For instance, briquetting of oxide has been proposed as a method of preventing dispersion during handling and storage. Though perhaps simple in design, practical and demonstrated processes to accomplish such remediation often do not exist. All processes and alternatives must be evaluated to assess their net impact on waste generation and other ES&H concerns.

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Consideration should also be given to the selection of process technologies for preparation of candidate forms. The level of waste generation varies substantially for process alternatives.

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Some disablement options generate more waste than others!!

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A severe waste impact is encountered for dilution to less than 0.5 at.% with non-nuclear material such as glass.

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4.3. Contingencies for Ultimate Disposition

A consideration of possible future issues and needs is essential, especially since the decision on final disposition remains unanswered. Plutonium removed from stockpile might be required for future weapon fabrication or used as an energy source. Alternatively, Pu might be diluted and packaged for long-term geologic disposition or monitored above-ground storage. Advantage is gained by choosing storage options that preserve maximum flexibility and provide minimum difficulty in responding to such future needs. Any option that causes problems with handling and reuse, or that causes reconstitution to be hazardous, difficult, or costly for a potential adversary will create similar problems for the custodian state.

The choice of storage form is a key factor in contingency planning. The decision is inexorably tied to the issue of waste generation. Excess weapons-grade plutonium exists almost entirely as pure metal in pits. Maintaining purity and metallic form minimizes waste generation now and in the future, while preserving flexibility for all currently suggested disposition options. Since the formation reactions of all chemical compounds from the metal are thermodynamically favored without the use of secondary reagents, the preparation of any final form is most efficiently accomplished through the use of metal. Interim storage of all plutonium compounds involves waste generation during their initial preparation for storage and during subsequent reprocessing to the final form. Separate technologies and equipment will most likely be required to prepare material for interim storage and for final disposition. The likelihood that interim and final forms will be identical is small unless a decision on ultimate disposition is made soon.

4.4 Transparency

Verification of nuclear material quantity and disposition without simultaneously divulging sensitive weapon-design or national-security information can be difficult. These problems are most acute for storage of intact or altered pits. In these cases, verification will likely rely on radiation signatures (neutron and gamma) that can be measured through the walls of a storage container or weapon case or on calorimetric measurements that can be made without disclosing component geometry. In order for such methods to be quantitative, information must be provided for the chemical and age-corrected isotopic composition of the nuclear material as well as for the relevant absorption or enhancement (α -n) characteristics of the storage configuration.

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Calorimetry is slow and the equipment is relatively expensive. Thermal imaging or acoustic resonance techniques may be effective but are not demonstrated.

Verification of pit disablement is both process and component specific.

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A combination of these methods could be employed to ensure against deliberate deception or falsification of a disablement process.

Transparency is more readily achieved for extracted material because the plutonium has neither a classified geometry nor is contained within other classified shapes. Visual inspection and mass verification are augmented by radiation measurements, isotopic analysis, and other destructive and nondestructive chemical methods. Complications arise in verifying that all plutonium originally present in a weapon is accounted for after extraction. Verification during the extraction process requires a balance of incoming and outgoing material and depends on the validity of the initial measurement and on the ability to accurately determine plutonium levels in process waste streams. A complication to plutonium inventory verification in dilute forms would be present because of difficulties in analysis.

5.0 CONCLUSIONS

The selection of a preferred technical option for plutonium storage is confounded by the complexity of technical issues. We have attempted to make a systematic evaluation of credible storage options and compare the relative merits of each with regard to several important issues. The results of this analysis are presented in Table I. Although the relative importance of individual issues is difficult to establish and subject to the reader's perspective, each of the issues listed significantly impacts decisions regarding storage options. All qualitative evaluations in Table I are on an absolute scale.

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Table 1. Evaluation of Plutonium Storage Options

Option	Direct Use Possible	Easily Reconstituted	Feasible	Development Required	Applicability	Waste Generation	ES&H Concerns	Flexibility
A. Intact	yes	yes	yes	minimal	all	none	few	high
B. Altered								
DoE b(3)	yes?	yes	yes	modest	some	minimal	some	moderate
2. Inert Material (many)	?	yes	yes	extensive	most	high	many	moderate
3. Reactive Material	?	yes	yes	modest	all	high	some	high
DoE b(3)	?	yes	yes	modest	most	minimal	many	high
	?	yes	yes	extensive	most	modest	many	limited
	?	yes	yes	extensive	most	modest	many	limited
	?	yes	yes	v. extensive	many	high	many	v. limited
C. Extracted								
1. Metal	yes	yes	yes	minimal	all - N/A	minimal	few	high
2. Oxide	yes	yes	yes	min or	"	modest	some	limited
3. Hydride	yes	yes	yes	min or	"	minimal	many	high
4. Nitride	yes	yes	yes	modest	"	modest	many	limited
5. Carbide	yes	yes	yes	extensive	"	modest	many?	limited
6. Alloys	yes?	yes	yes	extensive	"	high	many?	v. limited
7. Mixed Oxides	yes?	yes	yes	min or	"	high	few	v. limited
D. Extracted/Diluted								
1. Isotopic	yes	no	no	extensive	"	varies	few?	low
2. Radioactive (HLW)	no	no	no	v. extensive	"	high	excessive	none
3. Extreme Dilution	no	no	no	modest	"	v. high	many	minimal

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The issues examined in the first and second columns of Table I are particularly important for addressing proliferation and security concerns. The column labeled "direct-use possible" considers the possibility of using without modification the pit or altered pit in a nuclear weapon, and in the case of extracted material (section C and D) considers the possibility of using the stored, unaltered material in device fabrication. In the second column, "easily reconstituted" provides an evaluation of the difficulty in reprocessing the stored material to metallic plutonium.

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The potential
advantage of this option as a non-proliferation measure must be weighed against factors such as waste generation, ES&H concerns, and flexibility which appear in the right-hand columns of Table I. Verification of nuclear material content and detection of diversion or theft is extremely difficult with this storage form. Severely diminished flexibility is a consequence of extreme or high-level waste dilution because reprocessing to other

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material forms is difficult, if not prohibitive, from an ES&H standpoint. In addition, high-level waste or extreme dilution is appropriate only if these choices are acceptable options for final disposition.

The assessment of reuse and reconstitution options in Table I leads to several important conclusions. A magical solution does not exist for solving the proliferation problems associated with storage of plutonium. Neither alteration of pits nor conversion of metal to another material form can be justified on the basis of their effectiveness as a technical nonproliferation measure. An important conclusion is that safeguards and security are essential and provide the only available technical and physical means for addressing the proliferation risk of stored excess plutonium.

Since most options in Table I are feasible, a ranking of their suitability for interim storage rests with remaining issues in Table I. A survey of the assessments show that methods for pit alteration require substantial development, are not generally applicable to all pit types, generate substantial waste, and reduce flexibility. Performance in these categories is improved for the extracted storage options outlined in Section C of Table I. However, the most attractive option remains storage of excess plutonium as intact pits.

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The preceding conclusions are supported by prior assessment and by experience in

storage of various plutonium forms. In 1981, a DOE task force concluded that "metal is the best characterized, best understood, and easiest to store physical form of

plutonium."¹⁴⁸ The capability for storage of plutonium metal has been developed and

demonstrated during more than forty years of production and stockpile-storage of pits.

Techniques for preventing corrosion of the stored metal are well established. The

suitability of well-characterized metal as a storage form is evidenced by the remarkable

success of thousands of "pit tests" that span a period of over thirty years for some weapon

systems. Although experience in storage of weapons grade oxide is relatively limited, a

substantial database exists for storage of reactor-grade material. Large quantities of oxide

have been stored for up to ten years without incident. In these cases, the material was

prepared, certified and packaged according to well-defined procedures. Our conclusions

regarding preparing, certifying, and packaging of oxide for storage are consistent with the

findings of numerous groups.^{149,150,151} When the recommended procedures have not

been followed, failures and accidents have occurred. Indeed, unfavorable incidents have

occurred with both metal and oxide when preparation and packaging were inadequate.

6. RECOMMENDATIONS

We recommend that excess plutonium be stored as intact pits. Development activities

include definition of specifications for storage containers that accommodate all pit types.

Storage containers should immobilize tubulation, have certifiable closures, maintain a

safe storage temperature, and provide enhanced containment in the event of fire. Since

removal of tubulation destroys the certified pit closure, tube removal must be carefully

examined and may require additional development. Appropriate surveillance procedures

for monitoring behavior and changes during storage can be adapted from stockpile

evaluation procedures for similar pit types.

If extraction of plutonium is required for interim storage, we recommend that metal

be stored as critically safe castings. Implementation of this option will benefit from

development of advanced process technologies with the goal of minimizing waste and

reducing radiation exposures to personnel during recovery and casting. In addition,

specification and design of a storage container(s) must be completed and requisite

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procedures for metal preparation, certification, and packaging must be developed and implemented. Storage facility issues, surveillance requirements, and MC&A procedures must be established.

We recommend that all weapons-grade oxide and oxide-containing residues be stored in their existing material forms during the interim period. Container specifications must be established and methods for preparation, certification, and inventory of material from diverse sources must be developed. Surveillance procedures for oxide must also be defined and established. The suitability of storing various non-oxide residues and techniques for processing those materials to storable forms should also be investigated. If storage of extracted plutonium as metal is unacceptable due to non-technical considerations, we recommend that all plutonium be stored as oxide. Procedures must be developed to efficiently convert large quantities of metal to certified oxide with minimal waste generation.

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Our technical assessment and analysis of storage issues leads to general recommendations about packaging requirements and storage container design. Stored materials (intact pits, altered pits, extracted metal, oxide) must be well-characterized and certified. In the case of extracted materials, handling procedures must be established to ensure that integrity is preserved during packaging. Essential requirements for storage containers are strict exclusion of all reactive and radiolizable materials from the container (primarily organics and water), and isolation of stored material from the ambient atmosphere by one or more metal seals.

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Examination of reuse and reconstitution issues leads to general recommendations regarding reduction of proliferation risks.

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Safeguards and security remain the primary technical means to prevent diversion and use of stored plutonium by proliferant states and sub-national groups.

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